

Binding energy of localized biexcitons in quantum wells.

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Abstract

A variational calculation of the ground state energy of a biexciton in a GaAs/AlGaAs quantum well is presented. The well width fluctuations leading to trapping of the biexcitons are modeled by a parabolic potential. The results obtained for different well widths are compared with recent experimental data. Good agreement is obtained both for the biexciton binding energy and for the Haynes factor. We find that the structure of a biexciton is similar to the one of the H_2 molecule.

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In the last few years considerable experimental progress was made in detecting biexcitons in semiconductor systems. A biexciton is a system consisting of two excitons which are bound together. Since the first observation of biexcitons in quantum wells reported by R.C. Miller *et al.*, [1] there have been many studies, both experimental [1,3-8] and theoretical, [2,9,10] on this subject. Kleinman [2] developed a variational model that gives results in agreement with the first experimental [1,3] data. However later experimental studies, carried out with more advanced techniques, have reported substantial larger values for the binding energy [4-8] as compared to the early experiments. For example the Haynes factor, $f_H = E_b^{XX}/E_b^X$, which is the ratio between the biexciton binding energy (E_b^{XX}) and the exciton binding energy (E_b^X), found by Birkedal *et al.* [8] has a value in the range 0.19-0.22 for well widths between 80 and 160Å, while Kleinman predicts a value in the range 0.11-0.12. In order to explain this difference between theory and experiment, calculations were carried out with different techniques as well as with new assumptions [9] on the spatial form of the biexciton. Singh *et al.* [9] assumed a square-like arrangement of the electrons and the holes in a two dimensional biexciton which resulted in $f_H = 0.228$. The latter approach is rather *ad hoc* and does not include the finite thickness of the biexciton and consequently is not able to explain the well width dependence of the biexciton binding energy.

The aim of the present paper is to explain the recent experimental results by considering localization effects on the biexciton. This localization can be a consequence of the modulation of the thickness of the quantum well. Indeed for a quantum well of width L a variation in well width of ΔL produces a change in the zero point energy of the order of $\Delta L \hbar^2 \pi^2 / (m L^3)$. For a quantum well of 160Å a fluctuation of about 2.5Å induces a zero point fluctuation of the order of 0.5 meV which compares to a biexciton binding energy of about 1.5meV.

Using the effective mass approximation a biexciton in a quantum well can be described by the Hamiltonian

$$\hat{H}_{XX} = \hat{H}_{1X} + \hat{H}_{2X} + \sum_{i=e,h} (-1 + 2(\delta_{i,e} + \delta_{i,h})) \frac{e^2}{|\vec{r}_{1i} - \vec{r}_{2i}|} + V_{conf}(z) + \sum_{i=e,h} \sum_{j=1,2} \frac{1}{2} m_i \omega^2 \rho_{j,i}^2, \quad (1)$$

with

$$\hat{H}_{iX} = -\frac{\hbar^2}{2m_e^*} \nabla_{ie}^2 - \frac{\hbar^2}{2m_h^*} \nabla_{ih}^2 - \frac{e^2}{|\vec{r}_{ie} - \vec{r}_{ih}|}, \quad (2)$$

where the indexes 1, 2 indicate the first and second exciton, m_i^* is the effective mass of the particle i , and $V_{conf}(z)$ is the confining potential associated with the presence of the quantum well. ω is the frequency of the shallow parabolic confining potential in the quantum well plane that models the quantum well width fluctuations and $\vec{\rho}$ is the projection of \vec{r} in the plane orthogonal to the well axis. The confinement energy is much larger than the biexciton and exciton binding energy which allows us to treat the system as a quasi-two dimensional system, i.e. we can separate the contribution to the wave function along the quantum well axis, chosen as z-axis, from the contribution along the plane, the $\vec{\rho}$ -plane,

$$\Psi(\vec{r}_{1e}, \vec{r}_{2e}, \vec{r}_{1h}, \vec{r}_{2h}) = \mathcal{F}(z_1, z_2, z_a, z_b) \psi(\vec{\rho}_{1e}, \vec{\rho}_{2e}, \vec{\rho}_{1h}, \vec{\rho}_{2h}). \quad (3)$$

The component of the wave function along the z-axis is taken as a product of the 1D ground state wave functions for an electron (hole) in a hard wall quantum well. Averaging the Hamiltonian over the z-component $\mathcal{F}(z_{1e}, z_{2e}, z_{1h}, z_{2h}) = f_e(z_{1e})f_e(z_{2e})f_h(z_{1h})f_h(z_{2h})$ we obtain the following effective 2D Hamiltonian

$$\begin{aligned} \hat{H}_\rho = & \frac{1}{1+\sigma} (\sigma \Delta_a + \sigma \Delta_b + \Delta_1 + \Delta_2) - 2(U_{1,a} + U_{1,b} + U_{2,a} + U_{2,b} - U_{a,b} - U_{1,2}) \\ & + \frac{1}{4} (1+\sigma) \left[\frac{1}{\sigma} \omega^2 (\rho_a^2 + \rho_b^2 + \rho_1^2 + \rho_2^2) \right], \end{aligned} \quad (4)$$

where $\sigma = m_e/m_h$ is the mass ratio between the electron and the hole and $U_{i,j}$ is the effective Coulomb potential obtained by averaging the real Coulomb potential over the wave functions along the z-direction. In Eq.(4) we expressed the energy in units of $R_y = e^2/2\epsilon a_B$ and the length in units of $a_B = \epsilon \hbar^2/e^2 \mu$ with ϵ the static dielectric constant and μ the

in-plane reduced mass of the electron-hole system. Using $\sigma = 0.68$, i.e. $m_e/m_0 = 0.067$, $m_h/m_0 = 0.099$, $\epsilon = 12.1$ for a GaAs/AlGaAs with concentration of Al=25%, we find $R_y = 3.7meV$ and $a_B = 160\text{\AA}$.

It has been shown [11] that $U_{i,j}$ can be well approximated by $1/\sqrt{\lambda^2 + \rho^2}$, where $\lambda = 0.2L$ with L the width of the well which is valid for hard well confinement. The latter approach is a very good approximation for the wide quantum wells considered in the present paper. Using this approximation the Hamiltonian (4) was solved with the stochastic variational technique of Ref.12 with the trial wave function taken as a combination of correlated Gaussian functions,

$$\psi = \sum_{n=1}^K \Phi_{nLs}, \quad (5)$$

$$\Phi_{nLs} = \mathcal{A}\{\chi_{SMs} Y_{LM_L} (\sum_{i=1}^3 u_{ni} \vec{\zeta}_i) \exp(-\frac{1}{2} \sum_{i,j=1}^3 A_{nij} \vec{\zeta}_i \cdot \vec{\zeta}_j)\}, \quad (6)$$

where $\vec{\zeta}_1$ and $\vec{\zeta}_2$ are the distance vectors between the hole and the electron in the first and in the second exciton respectively and $\vec{\zeta}_3$ is the distance between the centers of mass of the two excitons, χ_{SMs} is the spin function, Y_{LM_L} is the angular function and \mathcal{A} is the antisymmetrization operator. The interaction among the different particles is taken into account via the non diagonal terms of the matrix \mathbf{A} . The best set of variational parameters $\{u_{ni}, A_{nij}\}$ is found using a stochastic method. The dimension of the basis K is increased until the required accuracy is achieved. Here we are only interested in the ground state, and consequently, the total angular momentum L and the total spin S are zero.

The biexciton binding energy is obtained as follows

$$E_b^{XX} = 2E^X - \tilde{E}^{XX} \quad (7)$$

where $\tilde{E}^{XX} = (E^{XX} - 4*\omega)$ is the biexciton ground state energy as referred to the four free particle in the shallow parabolic confinement potential and E^X the ground state of a *mobile* exciton.

The quantum well width fluctuations (ΔL) shift the zero point energy of the electrons and holes by $\Delta L \hbar^2 \pi^2 / (mL^3)$ and thus it is reasonable to assume that the shallow confinement ω

is inversely dependent on some power of the well width L . These considerations suggest to search for such a dependency in the form of $\omega(L) = a/L^n$. In order to do this we considered the experimental data reported in various experiments [4-8] and we used ω as a fitting parameter. The obtained confinement frequencies are plotted in Fig. 1. The influence of the confinement on the biexciton binding energy is shown in the inset of Fig. 1 for different values of the quantum well width. Note that E_b^{XX} increases almost linear with ω . On the basis of the above zero point energy argument we expect a $\omega \propto L^{-3}$ which seems to agree with the experimental results for $L/a_B < 0.7$. Noting that there is a lot of scatter between the different experimental results it seems that the best overall behaviour of ω is given by $\omega = 0.06/L^{-1}$, although a constant value of $\omega = 0.068R_y \approx 0.26meV$ also agrees with the experimental results, at least for $L/a_B > 0.6$

In Fig. 2 we plot our $\omega = 0$ biexciton binding energy are comparable to those found by Kleinman although our biexciton and exciton energy are considerably smaller. Adding a shallow confinement potential in the quantum well plane increases the biexciton binding energy substantially (about a factor of 2) and brings the theoretical results in agreement with the experimental results. We show our results for a constant confinement of $\hbar\omega = 0.068R_y$ and for a well width dependent confinement of $\hbar\omega/R_y = 0.06/(L/a_B)$. The latter gives a better overall agreement with the experimental data. The different experimental results are from different quantum wells which have been not grown under the same conditions and consequently the well width fluctuations can also be substantially different.

The Haynes factor, which is the ratio between the biexciton energy and the exciton energy, is found experimentally to be almost independent on the width of the well. Our theoretical results, see Fig. 3, seem to confirm this and leads to $f_H \approx .22$ which compares to the value, $f_H = 0.228$, found by Singh *et al.* Although our theoretical results show a weak well width dependence they fall within the scatter of the experimental results. weak well width dependence but

Note that the previous calculation by Kleinman results into $f_H = 0.13$ which is about a factor of two smaller than the one found experimentally and very close to the value we

found in the case $\omega = 0$.

In order to investigate the structure of the biexciton we evaluate the average distance between the two electrons, between the two holes and between the electron and hole which is defined as follows

$$\langle \rho_{ij} \rangle = \int |\psi(\vec{\rho}_{1e}, \vec{\rho}_{2e}, \vec{\rho}_{1h}, \vec{\rho}_{2h})|^2 |\vec{\rho}_i - \vec{\rho}_j| d\vec{\rho}_{1e} d\vec{\rho}_{2e} d\vec{\rho}_{1h} d\vec{\rho}_{2h}, \quad (8)$$

with $i, j = 1e, 2e, 1h, 2h$. The results are depicted in the inset of Fig. 4 as function of the well width. Note that the average electron-electron and the average hole-hole distances are comparable, and the average electron-hole distance is such that $\langle \rho_{eh} \rangle / \langle \rho_{ee} \rangle \approx 1.35$. For a square 2D biexciton as assumed by Singh *et al.* one has $\rho_{ee} = \rho_{hh} = \sqrt{2}\rho_{eh}$. Noticing that this equation is satisfied within 4% one may naively believe that the electrons and the holes are situated on the corner of a square.

Next we consider the pair correlation function for the electron-hole pair

$$P_{eh}(\rho) = \frac{1}{2} \sum_{i=1e, 2e} \sum_{j=1h, 2h} \langle \delta(|\vec{\rho}_i - \vec{\rho}_j| - \rho) \rangle, \quad (9)$$

and the one of the electron-electron (hole-hole) pair

$$P_{ee}(\rho) = \frac{1}{2} \sum_{i=\substack{1e, 2e \\ 1h, 2h}} \sum_{j=\substack{1e, 2e \\ 1h, 2h}} \langle \delta(|\vec{\rho}_i - \vec{\rho}_j| - \rho) \rangle, \quad (10)$$

which is plotted in Fig. 4. Note that the electron is much strongly correlated to the hole and that there is a high probability for the two particles to stay very close to each other. While electrons (holes) stay quite far from each other. This result argues against the model of a square biexciton proposed by Singh *et al.* and suggests that the electrons and holes orbit around each other like in single excitons and that the centers of mass of the two excitons are a certain distance apart which is approximately equal to the average hole-hole (electron-electron) distance. Such a configuration is similar to the one of a H_2 molecule.

In conclusion, we found that in order to explain the experimentally available results on the biexciton binding energy in quantum wells we have to assume that the biexcitons are trapped. The trapping potential is assumed to be parabolic which models the trapping

potential induced by the well width fluctuations found in real systems. Our results indicate that the trapping potential frequency has a smaller well width dependence than expected from a pure monolayer well width behaviour, except maybe for the quantum wells which are smaller than 100 Å. The Haynes factor is practically independent from the well width in agreement with the experimental results. By investigating the interparticle correlation functions we found that the biexciton can be considered like a H_2 molecule rather than a square arrangement of electrons and holes as proposed by Singh *et al.*.

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FIGURES

FIG. 1. The confinement trapping frequency as function of of the quantum well width L . The different symbols are the results obtained from the fitting to the experimental biexciton binding energy. The different curves shows the inverse power laws with $a=0.06$, $b=0.03$ and $c=0.02$ and L measured in Bohr radii. The inset shows the dependence of the biexciton binding energy on the confinement fo different well width.

FIG. 2. Comparison between different theoretical results for the binding energy of the biexciton (curves) and experimental data (symbols).

FIG. 3. The Haynes factor is plotted versus the well width. The dashed curve represents the results from the theory of Kleinman. The different symbols are the experimental results from different groups.

FIG. 4. The different pair correlation functions, for a biexciton in quantum well of width $L/a_B = 1$ and confinement energy $\hbar\omega/R_y = 0.068$, with $\rho_{ij} = |\vec{\rho}_i - \vec{\rho}_j|$. The inset shows the average distance between the different particles in the biexciton as function of the well width for a confinement energy $\hbar\omega/R_y = 0.068$.







